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**ATMOSPHERIC PARTICULATE MEASUREMENTS
IN NORFOLK, VIRGINIA**

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SUMMARY

Characterization of atmospheric particulates was conducted at a site near the center of Norfolk, Virginia. Air quality was measured in terms of atmospheric mass loading, particle size distribution, and particulate elemental composition for a period of 2 weeks. The objectives of this study were (1) to establish a mean level of air quality and deviations about this mean, (2) to ascertain diurnal changes or special events in air quality, and (3) to evaluate instrumentation and sampling schedules. Simultaneous measurements were made with the following instruments: a quartz crystal microbalance particulate monitor, a light-scattering multirange particle counter, a high-volume air sampler, and polycarbonate membrane filters. To assess the impact of meteorological conditions on air quality variations, continuous data on temperature, relative humidity, wind speed, and wind direction were recorded. Particulate elemental composition was obtained from neutron activation and scanning electron microscopy analyses of polycarbonate membrane filter samples. The measured average mass loading agrees reasonably well with the mass loadings determined by the Virginia State Air Pollution Control Board. There are consistent diurnal increases in atmospheric mass loading in the early morning and a sample time resolution of 1/2 hour seems necessary to detect most of the significant events.

INTRODUCTION

Langley Research Center, in cooperation with the Virginia State Air Pollution Control Board, Region VI, and Old Dominion University, made air quality measurements from August 6 to August 19, 1973, at a site near the center of Norfolk, Virginia. The selection of the site for the 2-week particulate sampling project was based on a request by the personnel from the Virginia State Air Pollution Control Board, Region VI. Utilizing instrumentation developed for aerospace research, Langley Research Center conducted an extensive, real-time characterization of atmospheric particulates. The

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objectives of this study were (1) to establish a mean level of air quality and deviations about this mean, (2) to ascertain diurnal changes, or special events in air quality, and (3) to evaluate instrumentation and sampling procedures. Air quality was measured in terms of (1) atmospheric mass loading, (2) particle size distribution, and (3) particulate elemental composition. Simultaneous measurements were made with the following instruments: a quartz crystal microbalance particulate monitor, a light-scattering multirange particle counter, a high-volume air sampler, and polycarbonate membrane filters. To assess the impact of meteorological conditions on air quality variations, continuous data on temperature, relative humidity, wind speed, and wind direction were recorded. Particulate elemental composition was obtained by neutron activation analysis and scanning electron microscopy of filter samples.

INSTRUMENTATION

The particulate sampling instrumentation used in this study is listed in table I and described in the following sections.

Light-Scattering Multirange Particle Counter

The light-scattering multirange particle counter (photometer), Royco Model 220, uses a photomultiplier to measure light scattered at a right angle by a single particle as it enters the illuminated sample volume. Errors caused by particle agglomeration were considered to be minimal under the sampling conditions of this study. The particle size range measurable with this instrument is approximately $0.5\ \mu\text{m}$ to $6.5\ \mu\text{m}$ in diameter, with this size range being divided into 100 size intervals by a pulse height analyzer (PHA). The PHA provides a size number distribution with the data recorded on paper tape by a high-speed printer and/or magnetic tape. Additional data, such as real time from a digital clock and various experimental parameters, also are recorded. Since the flow rate (2.8 liters/min) and sampling time are known, the size distribution and number of particles per cubic meter can be determined. By assuming a particulate mass density of $1.5\ \text{g/cm}^3$ (ref. 1), a plot of mass loading in g/m^3 as a function of time of day can be obtained for particles in the size range measured. The particle counting-data recording system was calibrated using a nebulizer-type aerosol generator with polystyrene latex spheres. Within limitations inherent in the light-scattering particle counters (ref. 2), the instrument yields time resolved, rapidly acquired, aerosol size distribution data. These data can be correlated with other measurements to detect short-term variations of particulate characteristics.

Quartz Crystal Microbalance Particulate Monitor

The quartz crystal microbalance particulate monitor (mass monitor), Celesco Model PM-39-D, is an impactor-type device (ref. 3) in which particulates are collected on an

oscillating quartz crystal and a rotating polycarbonate disk. The sensing crystal provides real-time data on mass loading, whereas the rotating disk collects material for subsequent elemental analysis. Located behind the sensing crystal is a second crystal which serves as a frequency reference and temperature compensator. The two crystals, in separate resonant circuits, produce a beat frequency which is increased when mass is added to one of the crystals. This beat frequency is converted to an analog voltage which is proportional to total mass collected. The analog signal is further differentiated to give a signal proportional to instantaneous mass loading. Both signals are scaled appropriately and recorded on a two-channel strip chart recorder directly in terms of total material collected in micrograms and instantaneous mass loading in micrograms per cubic meter. The size of particles collected on the crystal is in the range of $0.1\ \mu\text{m}$ to $10\ \mu\text{m}$ in diameter. Using scanning electron microscopy, pictorial and elemental identification can be made of particles collected on the polycarbonate disk. Both the sensing crystal and disk are coated with an adhesive to increase collection efficiency. This instrument can sample continuously in ambient levels of about $50\ \mu\text{g}/\text{m}^3$ for a period of 3 to 4 hours, at which time the crystal must be changed. Changing crystals requires approximately 1 minute.

High-Volume Air Sampler

The high-volume air sampler, MISCO Model 680, is essentially a high flow rate (850 to 1700 liters/min) pump with a head configuration for holding a 20- by 25-cm filter. The flow rate is obtained by measuring the pressure drop through the unit. There is also a timer for setting run times. Using 20- by 25-cm fiberglass filters, the particle collection efficiency exceeds 95 percent for particles $0.01\text{-}\mu\text{m}$ diameter and larger.

Polycarbonate Membrane Filters

The membrane filters, Nuclepore filters, are made from polycarbonate sheets approximately $10\ \mu\text{m}$ thick (ref. 4). The filters are made by boring holes with nuclear fission fragments and then enlarging the holes to desired size by placing the filters in an etching solution. This process yields a thin, uniform pore-sized, nonhygroscopic, transparent, uniform weight, collection surface for particulates. This clean, smooth filter material, relatively free of metallic impurities, lends itself very well to scanning electron microscopy and neutron activation analysis. The filter is clamped in a holder, and a vacuum pump is attached to the suction tube. Flow rates can range from 6 to 140 liters/min, depending on pore size, pump capacity, and other parameters. Particles are drawn to the filter surface and collected primarily by impaction, electrostatic forces, and physical adsorption. The particles collected can then be subjected to the analysis techniques described in this paper.

Weather Station

The Meteorology Research, Inc. (MRI) weather station measures and records the following meteorological parameters: wind speed, wind direction, temperature, and relative humidity. The station is a self-contained, battery-operated unit with accuracy of all measurements on the order of ± 2 percent. The chart drive is a clockwork mechanism with an accuracy of ± 60 seconds per 24 hours.

EXPERIMENTAL PROCEDURE

The instruments were installed in an air-conditioned mobile laboratory (fig. 1). In addition to the instrumentation, a Class 100 clean modular unit (bench), which permits filter loading and unloading on site, was also installed in the mobile laboratory.

The main sampling tube consisted of two sections of smooth-wall aluminum tubing, 2 meters long by 15 cm diameter. This 4-meter-long tube was connected by an O-ring seal to a feed-through port on the roof of the mobile lab, which placed the inlet of the main sampling tube at a height of 6 meters above the ground. Various diameter inlet tubes 25 cm long were contained in a circular flange and connected by an O-ring seal to the underside of the port. A blower in the port maintained a flow rate of approximately 1400 liters/min through the sampling tube. The inlet tubes were connected by short (30 to 60 cm) lengths of Tygon tubing (0.64 cm diameter) to the particle counter and the quartz crystal microbalance (mass monitor).

The light-scattering particle counter sampled particulates continuously, with a 10-minute sampling cycle. The data were checked periodically to determine if the particle concentration was large enough to cause coincidence effects (more than one particle in the scattering volume at any time) in the photometer. The counts seem to indicate that coincidence errors were not a significant problem. This procedure yielded continuous data on particle size distribution and concentration for the 2-week period, except for interruptions caused by minor instrument problems.

Due to limitations of the quartz crystal microbalance existing at the time of the experiment (which have subsequently been eliminated), a sample was collected for a 10-minute period once every hour from 0600 to 1800 hours. The sample periods were chosen to coincide with a 10-minute sample period of the light-scattering photometer.

The high-volume sampler was placed on a platform adjacent to and at the same height as the sampling tube on the mobile lab. The sampling was done on a 12-hour basis from 0600 to 1800 and 1800 to 0600 hours. The filters (collector and backing) were sheets of 20- by 25-cm fired glass fiber. The backup filter was used to prevent loss of collector filter weight due to loss of fibers and to prevent contamination by the sampler support screen.

The 47-mm-diameter polycarbonate membrane filters, in their holders, were mounted on a rack on top of the mobile lab. A vane pump was used to draw ambient air through the filters. The pump was connected to a sequential sampler and separate filters sampled for a 2-hour period during the 24 hours.

All filters, both the fiberglass and the polycarbonate membrane types, used during the test were subjected to the following procedure: The filters were placed in a large clean room, Class 100, with a controlled temperature of 22° C and a relative humidity of 40 percent, at the Langley Research Center, for 4 to 5 days prior to the initial weighing. The 20- by 25-cm filters were weighed on a microbalance (Mettler or Sartorius) and placed in a clean sealed envelope. The 47-mm-diameter polycarbonate membrane filters were weighed on a microbalance and placed in clean petri dishes. At the site, the filters were loaded and unloaded in the Class 100 clean modular unit (bench) within the mobile laboratory. After exposure, the filters were loaded into the various containers and returned to the large clean room at the Langley Research Center. They were exposed to the clean room environment for 4 to 5 days to equilibrate to preweighed relative humidity and were then reweighed.

The flow rate of the ambient air through the polycarbonate membrane filters was measured by using a dry gas volume meter with an accuracy of ± 2 percent. The flow rate was measured at the start and end of the 2-hour sample times. The average flow rate for the 2-hour samples was 27.4 liters/min.

ELEMENTAL ANALYSIS

Neutron activation analysis (NAA) is a very useful means of determining the elemental composition of many materials. (See ref. 5.) It is commonly used for analyzing particulate aerosols collected on substrates such as membrane filters. The basic principle of NAA is as follows: A sample of the material to be analyzed is irradiated with neutrons which interact with the atomic nuclei of the material. The atomic nuclei in the material become radioactive and emit various types of radiation as they decay. The energy spectrum of the gamma radiation emitted is characteristic of the elements present in the material. The intensity of the gamma radiation is proportional to the amount of the element present. The NAA detector can be calibrated with known quantities of each element; thus, it has the capability of determining quantity as well as elemental identity.

EXPERIMENTAL RESULTS

During the period of August 6 to 19, 1973, a large volume of data was collected on atmospheric mass loading, particle size distribution, particulate elemental composition,

and meteorological conditions by a variety of instruments. Approximately 150 000 data points per day were accumulated on particle size distribution. Reduction of such a volume of data involved averaging and plotting as a function of day and time of day for each day. The time of day plots revealed special events which were further analyzed in terms of frequency of occurrence and change in particle distribution, elemental composition, and meteorological conditions.

The atmospheric mass loadings (for the period of August 6 to 19, 1973) as determined from the 12-hour high-volume sampler filters and the 2-hour polycarbonate membrane filters are presented in figure 2. The 2-week mean mass loading, as determined with the high-volume sampler was $79 \mu\text{g}/\text{m}^3$ with the extreme values ranging from 40 to $192 \mu\text{g}/\text{m}^3$. The variations in the 2-hour polycarbonate membrane filter mass loading over the same 2-week period was from 6 to $382 \mu\text{g}/\text{m}^3$. The structure of this variation is illustrated in figure 2.

Particle number densities and size distributions, obtained by the multirange particle counter, were examined from three perspectives. First, each contiguous size distribution, for the range 0.5 to $6.5 \mu\text{m}$ in diameter, was viewed on an isometric plot for the time frame midnight to midnight. Data for a typical day, August 18, 1973, are presented in figure 3. Second, an integration over each size distribution was made to obtain total particle number density. These number densities, in the time frame midnight to midnight, for two representative days, August 15 and August 18, 1973, are shown in figure 4. Third, the size distribution data were least-squares fitted to the Junge power-law distribution. (See ref. 6.) According to such a distribution the larger the exponent obtained, the larger the number of small particles. The resulting exponent, Junge slope, is plotted as a function of the time of day for August 15 and August 18 in figure 5.

Meteorological data (surface temperature, relative humidity, wind speed, and wind direction) were reduced from continuous strip chart recordings by simple averaging over each hour. Data for August 15 and August 18, 1973, are shown in figure 6. Each data point shown is the midpoint for each hourly average.

Twenty-three polycarbonate membrane samples from 6 days were analyzed by neutron activation technique at Virginia Polytechnic Institute and State University. The system is capable of identifying 65 elements (11 being the lowest detectable atomic number). Several of the more abundant elements are shown as a function of time of day for August 15, 1973, in figure 7. The total amount of identifiable material represented approximately 24 percent by weight of the total material collected on the polycarbonate membrane filters.

Figure 8 shows the particulate mass loading as a function of time of day over a 24-hour period for August 16, 1973. The measurements were made with four different instruments, as indicated. The particle counter data were continuous with a time resolution

of 10 minutes. The mass monitor sampled for 10 minutes once each hour during the day, typically from 6:00 a.m. to 6:00 p.m. Data from each instrument represent an average over the 10-minute period. Each polycarbonate membrane filter collected for a 2-hour period. The data points are plotted at the midpoints of the sampling period. The high-volume sampler sampled for 12-hour periods, from 0600 to 1800 and from 1800 to 0600.

DISCUSSION OF RESULTS

In reducing the filter data, a problem was discovered in the microbalance. Because of mechanical problems, random errors were present. After repairing the microbalance and reweighing the control filters, the random error was determined to be $\pm 70 \mu\text{g}$. For flow rates associated with polycarbonate membrane filters and a sample time of 2 hours, the mass loading error is $\pm 21 \mu\text{g}/\text{m}^3$; for 12 hours, $\pm 3.5 \mu\text{g}/\text{m}^3$. For the flow rates associated with a high-volume sampler filter and a 12-hour sample time, the error is $\pm 0.07 \mu\text{g}/\text{m}^3$.

In conducting any experiment in which a sample is drawn from a much larger population, one is always concerned about the data being truly representative. For the 2-week period of this project, the mean atmospheric mass loading (determined by continuous 12-hour high-volume samples) was $79 \mu\text{g}/\text{m}^3$. The mean mass loading, determined by the Virginia State Air Pollution Control Board, Region VI, was $69 \mu\text{g}/\text{m}^3$ for this quarter, and the mean value was $88 \mu\text{g}/\text{m}^3$ for the previous year. (See ref. 7.) Thus, to a first approximation, a satisfactory agreement exists between the two separate sets of data.

The continuous light-scattering data, with a time resolution of 10 minutes, reveal a consistent positive event in particulate number density in the early morning. Figure 3 shows a good example of such an event. Examination of two representative days – August 15 (figs. 4(a), 5(a), and 6(a)) and August 18 (figs. 4(b), 5(b), and 6(b)) – shows no regular relationship between the early morning event and a change in size distribution or wind direction. On August 15, an event occurred in particle number density (fig. 4(a)) between 0600 and 1000 hours with little change in size distribution (fig. 5(a)) or local weather conditions (fig. 6(a)). In contrast, on August 18, an event between 0600 and 0800 (fig. 4(b)) was accompanied by a change in size distribution (fig. 5(b)). The winds on these 2 days were in opposite directions, coming from the southeast on August 15 (fig. 6(a)) and from the northwest on August 18 (fig. 7(b)). This tends to rule out the possibility of the particles coming from a single location. Since the event occurred in the early morning, the contribution of automobile traffic to the event was considered. If, however, automobile traffic was the source, this same kind of event should have occurred during the later afternoon traffic peak. This was not observed. Earl C. Kindle, Old Dominion University, suggested that this early morning diurnal occurrence may be

caused by a fumigation meteorological process. That process is due to a temperature inversion layer established at night, trapping particles and gases in it. At sunrise, the temperature profile rapidly changes and, consequently, a mixing occurs which conveys pollutants from the inversion layer to the ground level.

Further examination of 87 isolated events (table II) shows little overall correlation with wind direction and only minor correlation with size distribution changes. However, correlations do occur for certain events. Such an event occurred at 0100 on August 15 with changes in particle size distribution and number density (figs. 4(a) and 5(a)), which correlate with a wind direction change of approximately 180° (fig. 6(a)).

The elemental composition for August 15 (fig. 7) was examined for correlations. At about 0800, the time of the early morning event, the elements Na, Cl, Al, and Ti are increasing in mass loading. During this time period, the wind was from the southwest (fig. 6(a)). This wind direction would explain the increase in Na and Cl if they are assumed to be from sea salt. The sources of Ti and Al are unknown. Between 1100 and 1500, there is a decreasing trend in the concentrations for most of the elements plotted. This trend corresponds to the relatively low particle number density during this time period (fig. 4(a)). After 1500, there is a sharp increase in Fe with a slight increase in Ti. This increase occurs at about the same time as a number density increase which peaks at 2100 (fig. 4(a)) and also with a peak in polycarbonate membrane filter mass loading as seen in figure 2. While no definite conclusion can be made from a single day's data, some moderate correlations are apparent. The correlations with wind direction suggest the possibility of using this method for locating sources containing some of these elements.

A comparison of the results from different instruments utilized in this project and the different sampling times is shown in figure 8 for August 16. In general, trends in data agree for the different instruments. However, these instruments measure different size ranges of particles, have different collection efficiencies, and should not be expected to agree completely. In going from the 10-minute time resolution of the particle counter data to the 12-hour resolution of the high-volume sampler data, the increasing loss of information becomes quite apparent. Although the longer sample time is advantageous in smoothing random variations which may be of no interest, events which are of interest and have been shown as diurnally occurring are lost. From these data, it would appear that a sample time of $1/2$ hour should be sufficient to resolve events of interest.

CONCLUDING REMARKS

During the 2-week period in which air quality measurements were made at one site in Norfolk, Virginia, a set of instruments measuring the mass loading, particle size distribution, and elemental composition of the aerosols produced data requested by the

Virginia State Air Pollution Control Board, Region VI. Even though this study was conducted at a single location, the data obtained reveal the following useful correlations:

(1) The average mass loading of the air determined during this 2-week study was $79 \mu\text{g}/\text{m}^3$. This compares reasonably well with the State-determined average of $69 \mu\text{g}/\text{m}^3$ for the third quarter of the year.

(2) The data from polycarbonate membrane filters collecting aerosols for 2-hour periods were consistent with high-volume sampler collections. The polycarbonate membrane filters, however, gave a better indication of the variation of mass loading during the day and can be used to collect material for elemental analysis relatively easily and cheaply.

(3) The light-scattering multirange particle counter and quartz crystal microbalance particulate monitor gave consistent results. These results indicate that short-term (event-type) variations in the atmospheric mass loading can sometimes be missed by the 2-hour polycarbonate membrane filter averaging periods. A time resolution of $1/2$ hour seems necessary to detect such significant events.

(4) Many excursions from the ambient values were observed in atmospheric mass loading during this 2-week period. Most of these excursions appeared random with respect to a given time frame, wind direction, or changes in size distribution. Individual, isolated events may, however, be traced to particular sources by wind direction or size distribution. Several events did occur in a diurnal fashion; an excursion in mass loading consistently appeared in the early morning after sunrise. This occurrence is attributable to a fumigation meteorological process. Also, excursions frequently occurred around midnight. However, these consistent events do not always appear to be connected with wind direction and size distribution variations. Increased time resolution of the parameters gives new insight into the nature of the particulates and the factors which characterize and influence them.

Although neutron activation analysis can provide valuable information at a relatively low cost per sample, it does not yield all the information desired. Some of the elements that are not identifiable by this method are of interest in pollution studies. In particular, the elements with atomic numbers below 11 (such as carbon) and some heavier elements (such as lead) are not identified.

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TABLE I.- PARTICLE SAMPLING INSTRUMENTATION CHARACTERISTICS

Instrument	Type	Particle size (diam.) range, μm	Flow rate, liters/min	Type of data obtained
Multirange particle counter	Light scattering at right angle	0.5 to 6.5	2.8	Numbers of particles, size distribution, real time
Particulate monitor	Quartz crystal microbalance	0.1 to 10	1.8	Mass ($\mu\text{g}/\text{m}^3$), elemental composition, real time
High-volume air sample	High volume	All above 0.01	1133 to 1700	Mass ($\mu\text{g}/\text{m}^3$), elemental composition
Polycarbonate membrane filter	Filter media	All above 0.1	28.3 to 56.6	Size distribution, mass ($\mu\text{g}/\text{m}^3$) or particle count elemental composition

TABLE II.- EVENT OCCURRENCES AT WIND DIRECTION
AND CHANGES IN SIZE DISTRIBUTION^a

Excursions	Number of occurrences for wind direction ^b of -							
	N	NE	E	SE	S	SW	W	NW
Positive	11	1	2	11	10	5	1	3
Negative	11	1	2	8	12	3	2	2

Excursions	Number of occurrences for size distribution -		
	To larger particles	To smaller particles	For no change
Positive	10	11	19
Negative	8	8	20

^aEvent defined as excursion of mass loading.^bWind direction averaged over 1 hour.



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Figure 1.- Instrumentation van.

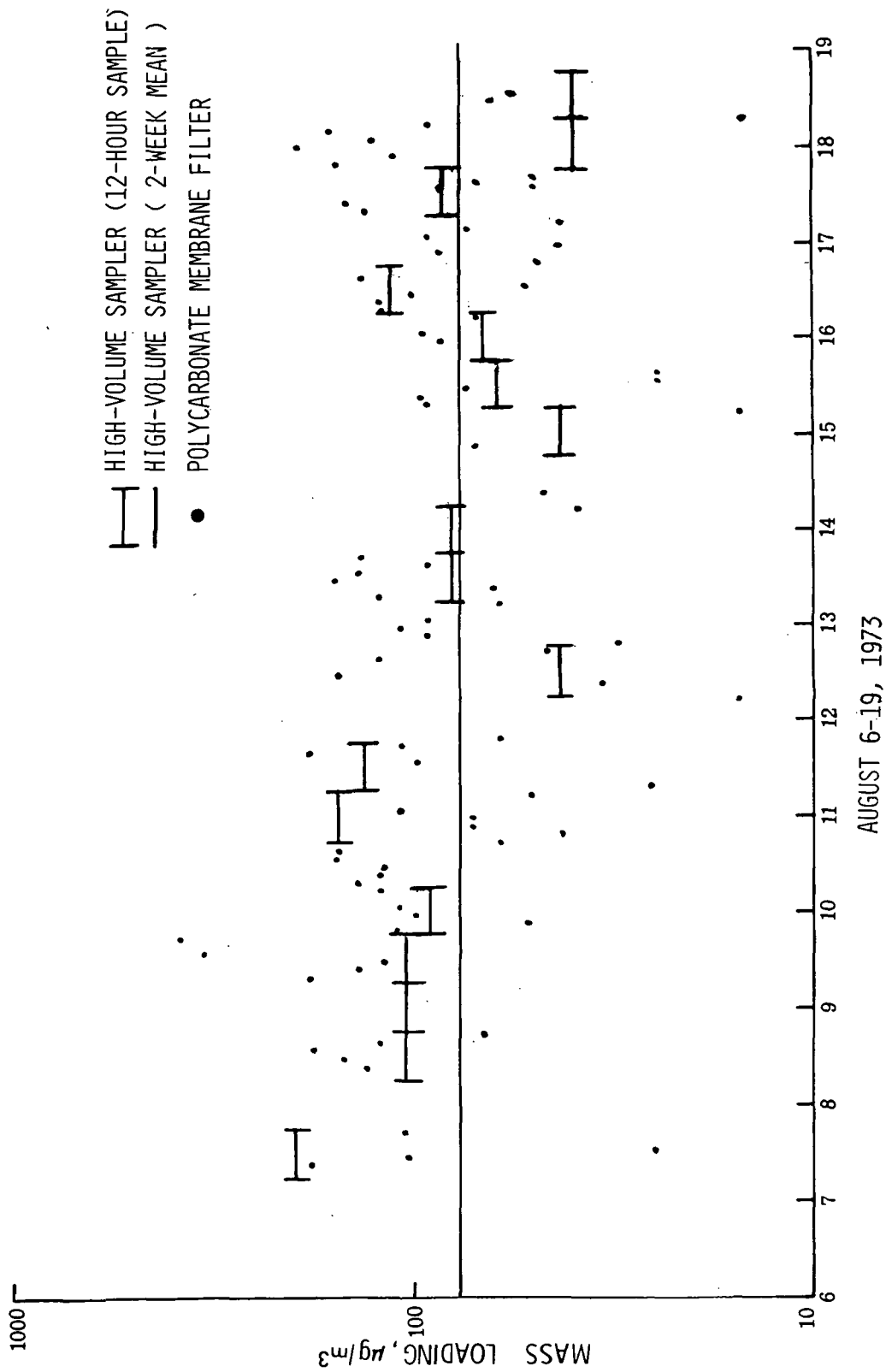


Figure 2.- Variations in atmospheric mass loading for August 6-19, 1973.

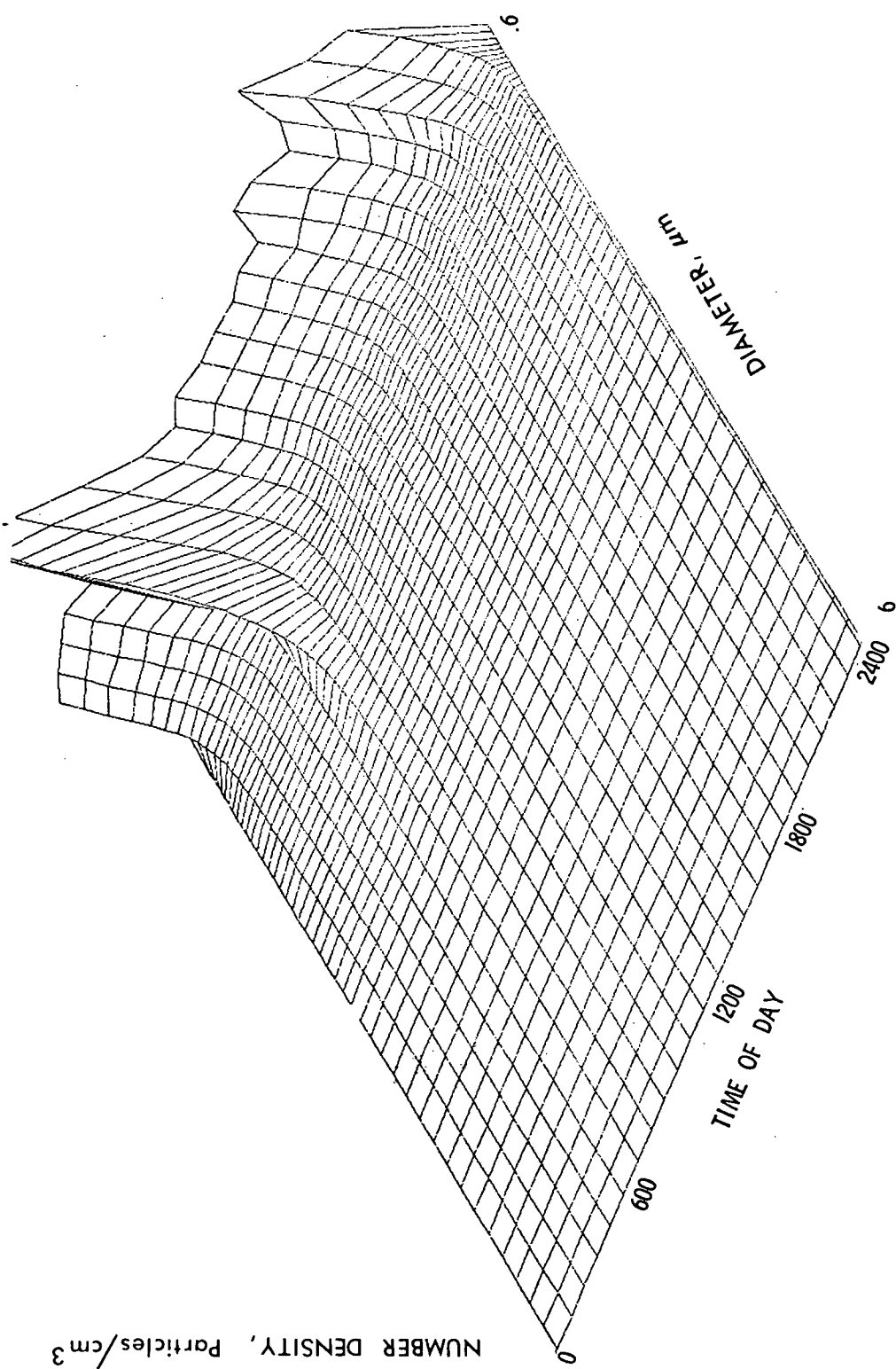
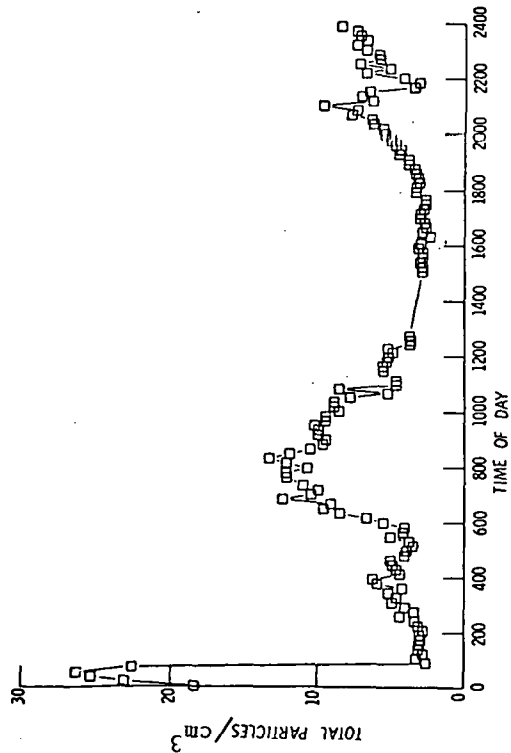
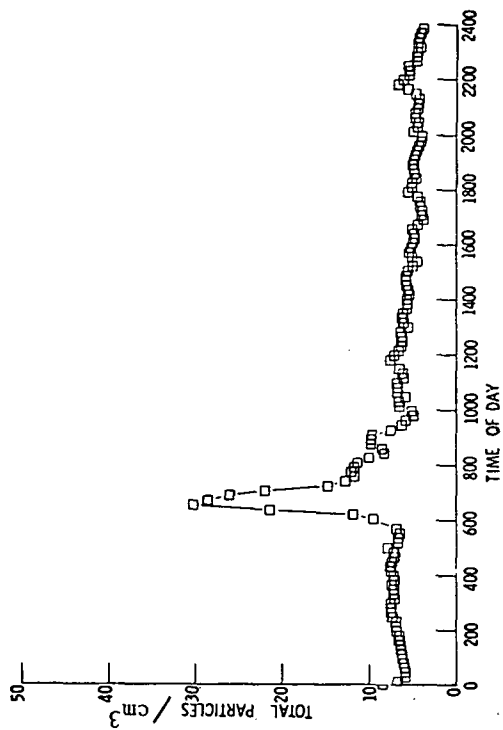


Figure 3.- Particle size distribution for August 18, 1973.

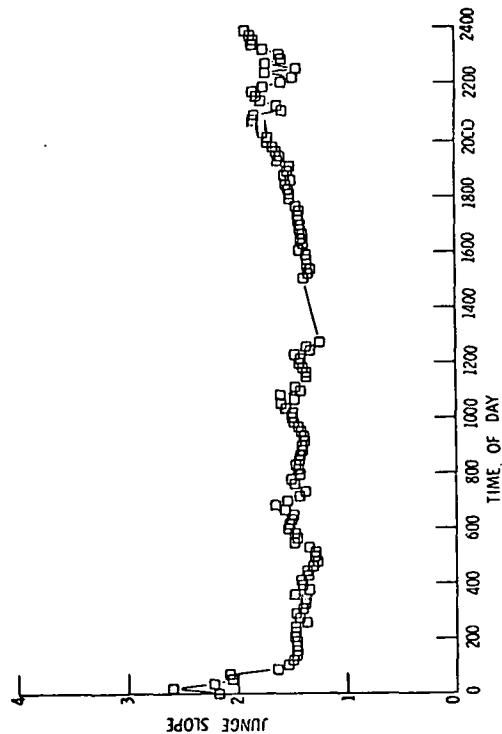


(a) August 15, 1973.

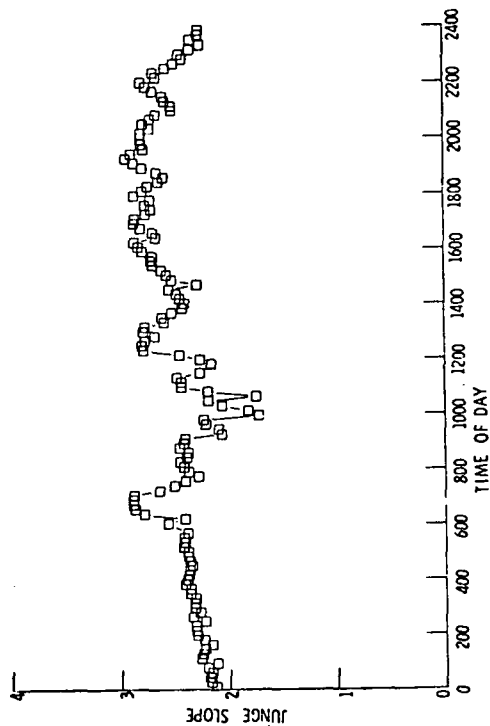
Figure 4.- Particle number density.



(b) August 18, 1973.

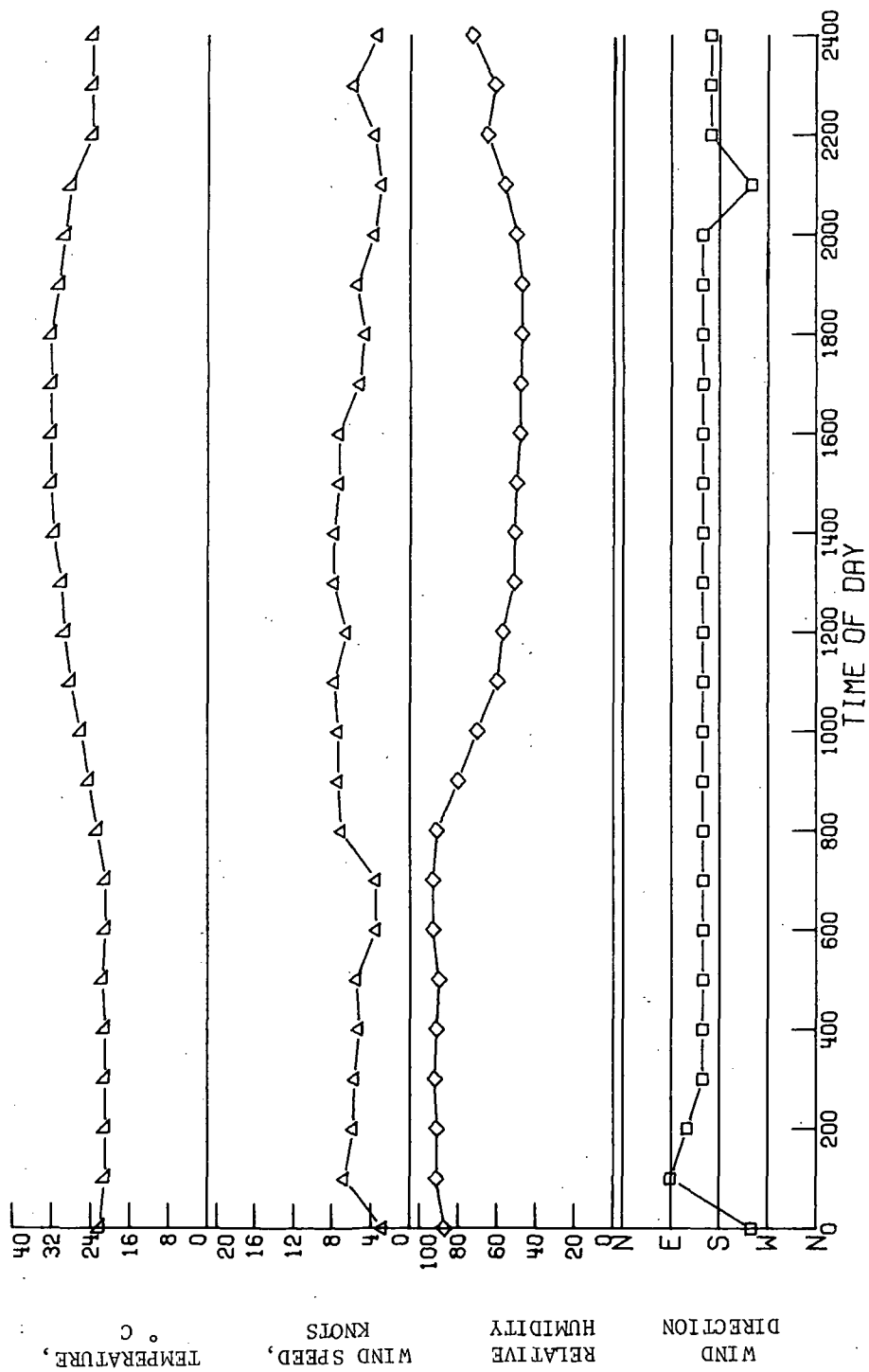


(a) August 15, 1973.



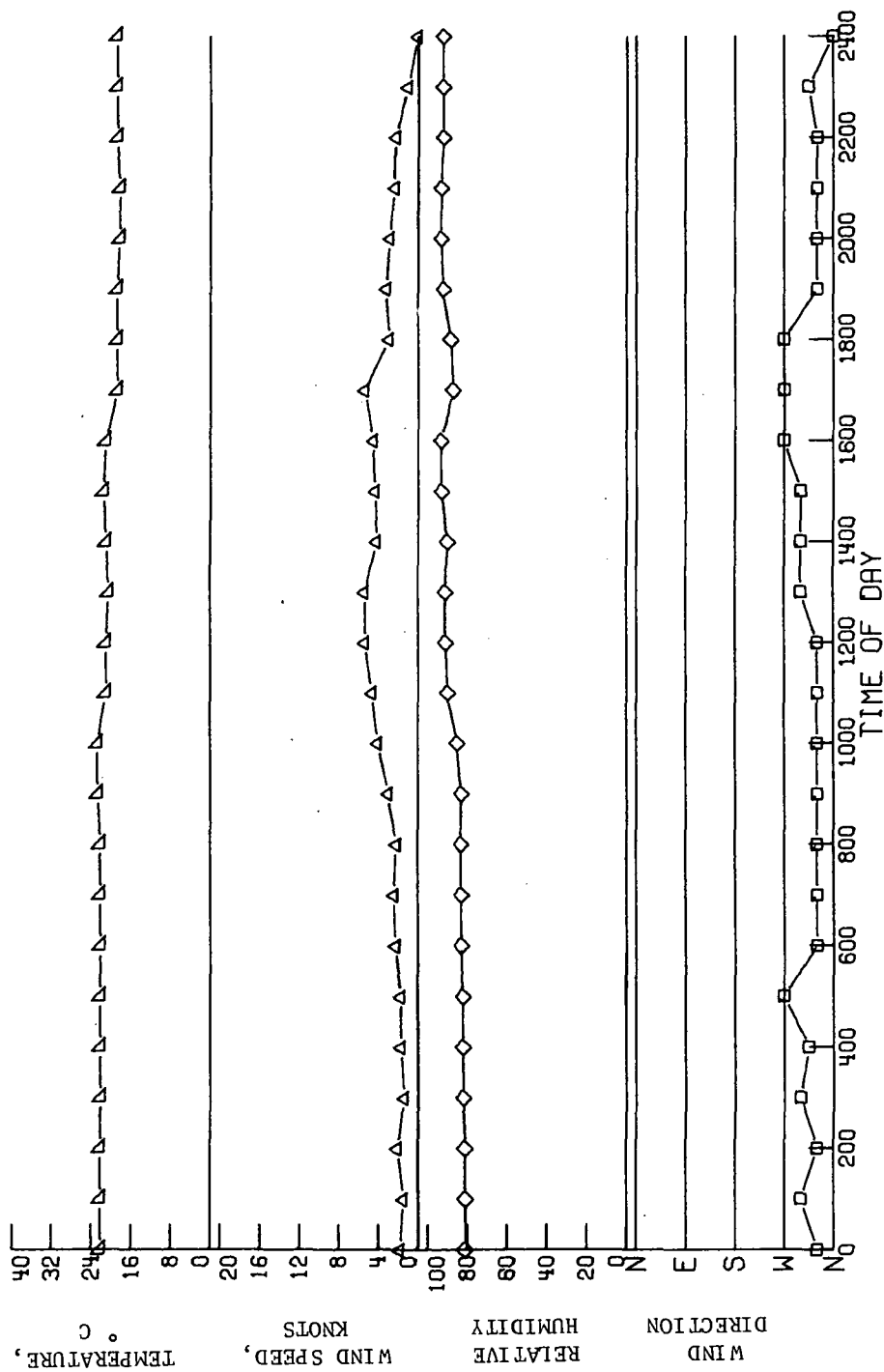
(b) August 18, 1973.

Figure 5.- Particle size distribution parameter.



(a) August 15, 1973.

Figure 6.- Meteorological data.



(b) August 18, 1973.

Figure 6.- Concluded.

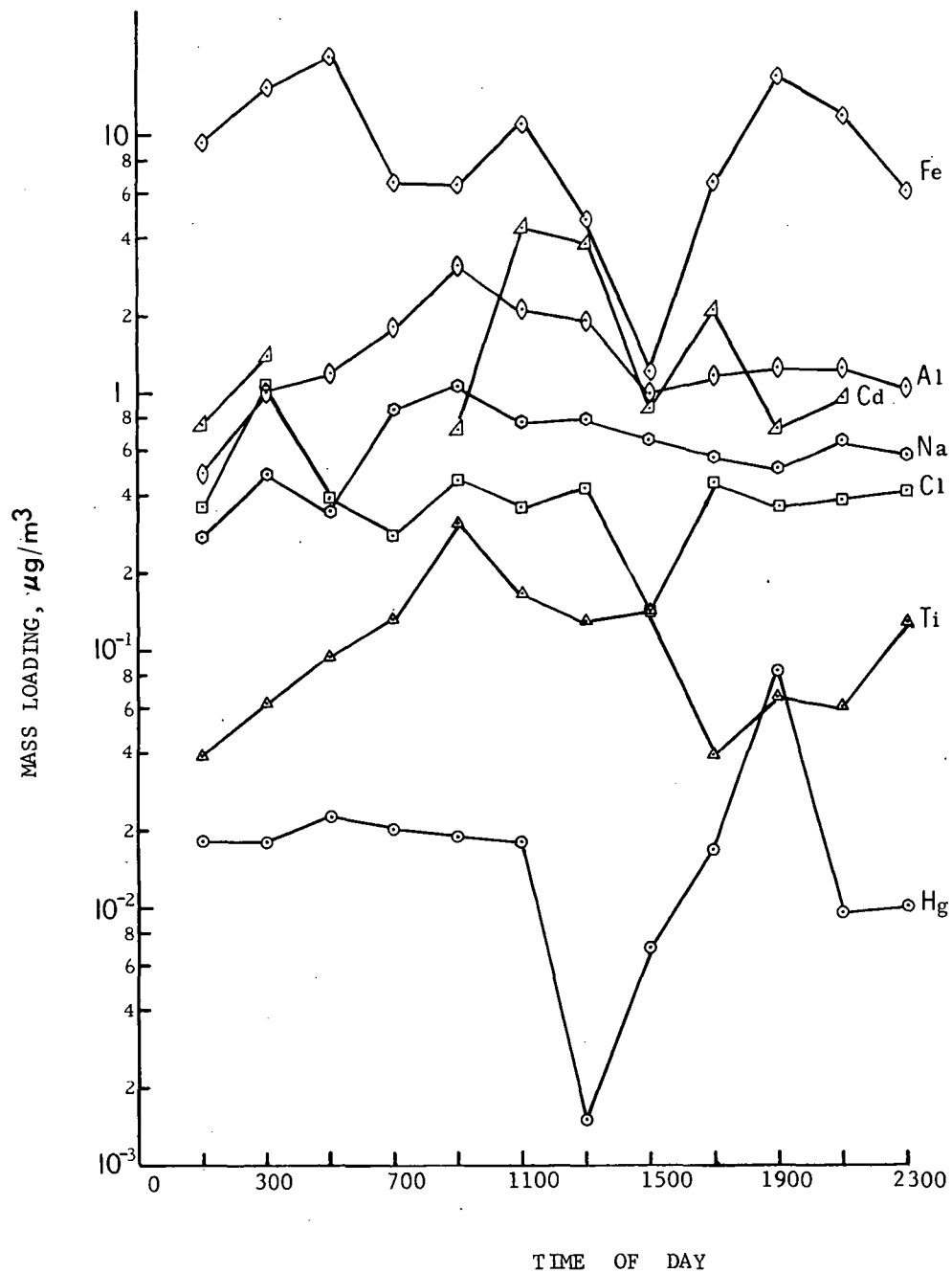


Figure 7.- Elemental atmospheric mass loading for August 15, 1973.

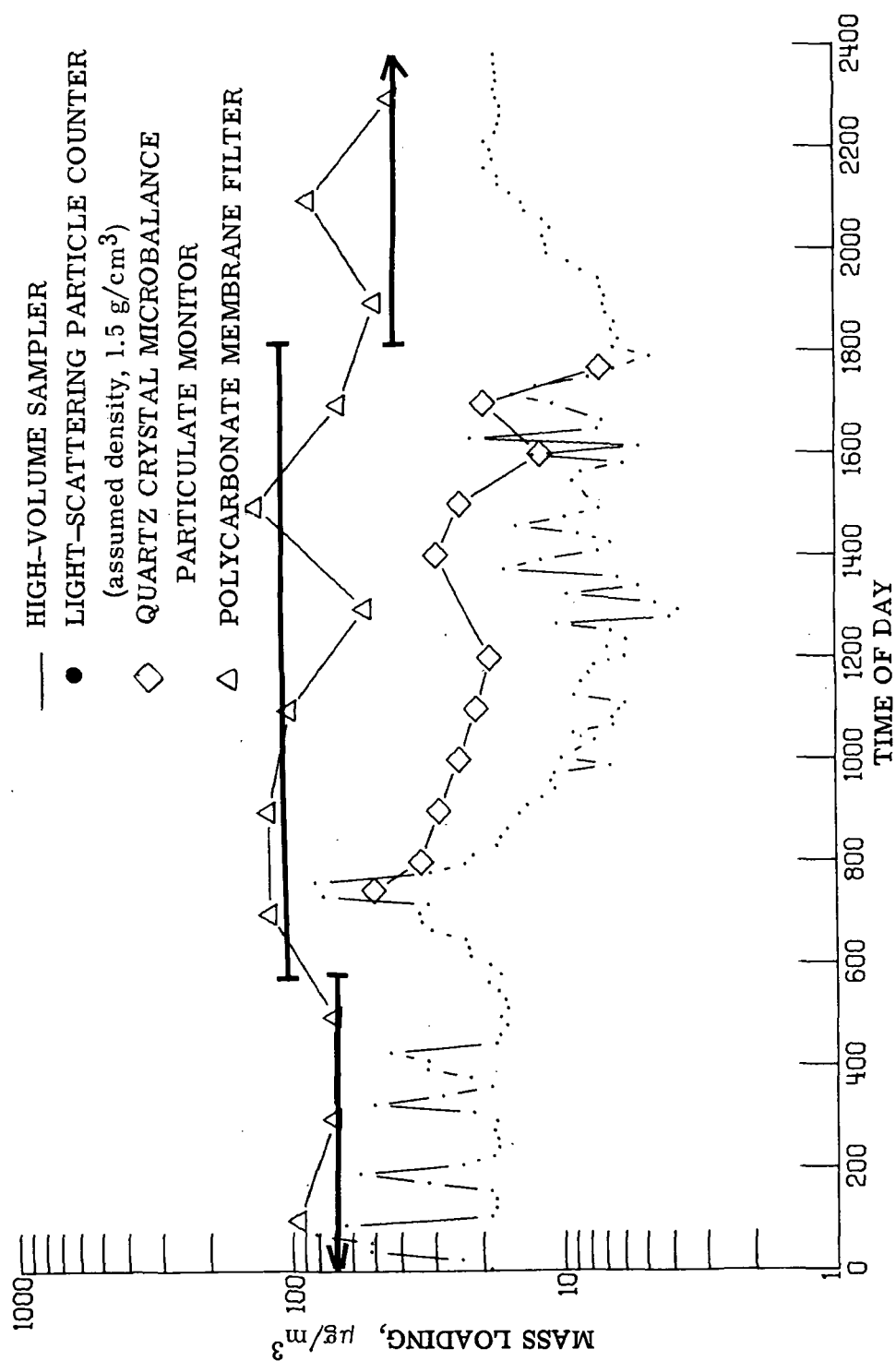


Figure 8.- Atmospheric mass loading for August 16, 1973.



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